Magnetic field-tuned quantum critical point in $CeAuSb_2$

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Transport, magnetic and thermal properties at high magnetic fields (H) and low temperatures (T) of the heavy fermion compound $CeAuSb_2$ are reported. At $H = 0$ this layered system exhibits antiferromagnetic order below $T_N = 6$ K. Applying B along the inter-plane direction, leads to a continuous suppression of T_N and a quantum critical point at $H_c \simeq 5.4$ T. Although it exhibits Fermi liquid behavior within the Néel phase, in the paramagnetic state the fluctuations associated with H_c give rise to unconventional behavior in the resistivity (sub-linear in T) and to a $T \ln T$ dependence in the magnetic contribution to the specific heat. For $H > H_c$ and low T the electrical resistivity exhibits an unusual T^3-dependence.

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When the long-range order is suppressed to zero temperature by tuning an external variable, such as pressure, chemical composition or magnetic field H, the system is said to exhibit a quantum critical point (QCP)[1, 2]. The magnetic field is an ideal control parameter, since it can be reversibly and continuously tuned towards the QCP. Two compounds with field-tuned QCP, $YbRh_2Si_2$ and $Sr_3Ru_2O_7$, reached prominence due to the non-Fermi liquid (NFL) behavior triggered by the quantum fluctuations associated with the QCP. In this letter we present a Ce-compound, $CeAuSb_2$, exhibiting a field-tuned QCP and unusual transport and thermodynamic properties. All three systems have a field-tuned QCP as a common thread, yet their behavior in high fields and low T are considerably different.

At zero field $YbRh_2Si_2$ exhibits a second-order phase transition into an antiferromagnetic (AF) state at $T_N = 70$ mK [3]. A magnetic field applied along the inter-plane direction drives T_N to zero at a critical $H_c \simeq 0.66$ T, leading to NFL behavior, i.e., a logarithmic increase of $C_e(T)/T$ and a quasilinear T dependence of the electrical resistivity ρ below 10 K [4]. Above H_c Fermi liquid (FL) behavior is recovered ($\rho \propto AT^2$ and constant Sommerfeld coefficient γ), with $A(H)$ and $\gamma(H)^2$ displaying a $1/(H - H_c)$ divergence as $H \rightarrow H_c$ [5]. A similar trend was recently found in YbAgGe [6].

Field-tuned anomalous metallic behavior in the vicinity of metamagnetism (MM) was studied in detail in $Sr_3Ru_2O_7$ [7]. At a MM transition the magnetization M increases rapidly over a narrow range of fields. The transition is of first order, since there is no broken symmetry involved, and terminates in a critical "end" point ($H^\star, T^\star$) [8]. In the anisotropic MM transition of $Sr_3Ru_2O_7$, $T^\star$ is found to decrease continuously as H is rotated towards the inter-plane c-axis [9], thus opening the possibility of a QCP in a first order transition [7, 8]. This scenario is supported by the T-linear dependence in ρ [7], a divergence of the coefficient A of the resistivity [7], the enhancement of the effective mass of the quasiparticles [10], and the $\ln T$-dependence of the specific heat γ [11]. Remarkably, at very low T and very close to the critical field H_c, ρ displayed a T^3-dependence [7]. The QCP of a MM transition is also believed to cause the rich phase diagram of URu_2Si_2 at high fields [12].

Among Ce compounds, so far there is evidence for a field-tuned QCP only in $CeCoIn_5$ [13] and $CeIrIn_5$ [14]. In these systems the QCP is believed to give rise to a (possibly unconventional) superconducting (SC) phase, in addition to NFL behavior. In $CeCoIn_5$ the nature of the magnetic correlations at low T is still unclear, in part due to the close proximity to SC, while in $CeIrIn_5$ the quantum critical behavior close to the metamagnetic (MM) transition is still under investigation.

In this letter we report on the anomalous properties of $CeAuSb_2$. $CeAuSb_2$ is a tetragonal metallic compound, which at $H = 0$ orders AF [15] with $T_N = 6.0$ K [16]. For $T < T_N$, $\rho(T)$ has the AT^2 dependence typical of a FL and the extrapolation of C_e/T to $T = 0$ yields a Sommerfeld coefficient of $\gamma \sim 0.1$ J/mol.K^2. Hence, $CeAuSb_2$ can be considered a system of relatively light heavy-fermions. Above T_N, on the other hand, $\rho(T)$ displays a T^α dependence with $\alpha \lesssim 1$ and, C_e/T has a $-\ln T$ dependence, both characteristic of NFL behavior due to a nearby QCP. A magnetic field along the inter-plane direction leads to two subsequent metamagnetic transitions and the concomitant *continuous* suppression of T_N to $T = 0$ at $H_c = 5.3 \pm 0.2$ T. As the AF phase boundary is approached from the paramagnetic (PM) phase, γ is enhanced and the A coefficient of the resistivity diverges as $(H - H_c)^{-1}$. When T is lowered for $H \sim H_c$, the T-dependence of ρ is sub-linear and the one of C_e/T is approximately $-\ln T$. These observations suggest the existence of a field-induced QCP at H_c. At higher fields an unconventional T^3-dependence emerges in ρ and becomes more prominent as H increases, suggesting that the FL state is *not* recovered for $H \gg H_c$.

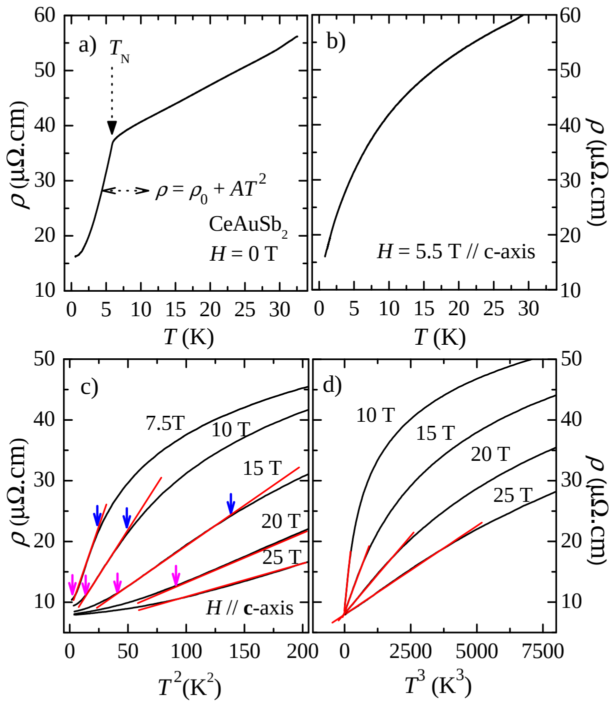

FIG. 1: a) T-dependence of the in-plane resistivity ρ of a CeAuSb$_2$ single crystal. At T_N a pronounced change in slope is observed, below which FL behavior is recovered. b) This behavior is suppressed and a sub-linear T-dependence is observed when $T_N \to 0$. c) At higher fields a T^2-dependence is obtained but only over a limited range of T as indicated by the vertical arrows (red lines are linear fits to a T^2-dependence). d) At the lowest T and largest H a T^3-dependence is observed (red lines are linear fits to a T^3-dependence)

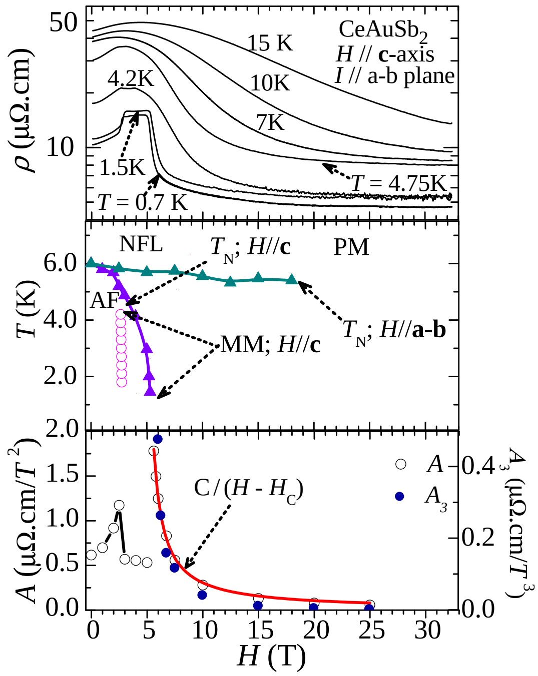

FIG. 2: Upper panel: In-plane resistivity ρ of CeAuSb$_2$ as a function of H. Middle panel: The resulting phase diagram indicating the AF phase boundary, the PM region, and the region in field where NFL behavior is found. Arrows indicate the critical fields of the metamagnetic transitions. Note that the effect of H on the Néel temperature T_N is very anisotropic. Lower panel: Coefficients A (open circles) and A_3 (blue circles) of the resistivity as a function of H. See text for details.

Single crystals of CeAuSb$_2$ were grown by the self-flux method, as described in Ref. [17], using high purity starting materials with excess Sb as a flux. Microprobe analysis confirms the stoichiometric composition of the samples as well as the absence of sub-phases. Electrical transport measurements were performed with the Lock-In technique in both, a Bitter and a superconducting magnet, coupled to cryogenic facilities . The magnetization as a function of T and H was obtained in a Quantum Design DC SQUID, as well as with a high field vibrating sample magnetometer. The heat capacity was measured in a Quantum Design PPMS system using the relaxation time technique. The heat capacity measurements were extended to higher temperatures in order to resolve the crystalline electric field (CEF) scheme. A Schottky-like peak centered around $T \simeq 50$ K was observed, suggesting an excitation energy of $\Delta = 110$ K between the ground and first excited doublets.

Figure 1a) displays the in-plane electrical resistivity ρ as function of T for $H = 0$ T. The vertical arrow indicates the onset of AF order, below which a T^2-dependence of ρ is obtained. Above T_N a NFL-like T^α-dependence with $\alpha \lesssim 1$ is observed. As H increases the AF order is gradually suppressed and in the vicinity of the AF to PM phase boundary a linear dependence on T emerges (see Fig. 1b)). At higher fields FL behavior, i.e., $\rho = \rho_0 + AT^2$, is found over a limited range of T as indicated in Fig. 1c) by the vertical arrows. The red lines are least square fits to the T^2-dependence. The slope, given by the A coefficient, decreases as H increases. At very low T and very high H ρ displays an anomalous T^3-dependence (see Fig. 1d)), suggesting, on the one hand, an unusual scattering mechanism, and, on the other hand, that the T^2-dependence may just be a crossover regime between T^α with $\alpha < 1$ and the T^3 regions.

The H dependence of ρ sheds more light on the role of the magnetic fluctuations (see upper panel of Fig. 2). For $T > 6$ K, a crossover from positive to negative magnetoresistance is seen in $\rho(H)$ at $H \sim 5$ T. As T is lowered below 6 K, two MM transitions emerge, with negative magnetoresistance developing at the AF to PM transition. The magnetic field suppresses the AF spin-correlations, giving rise to a gradual alignment of the spins and reducing this way the spin-flip scattering. The mechanism of the anomalous T^3-dependence in $\rho(H)$ is believed to be related to the alignment of the Ce-spins.

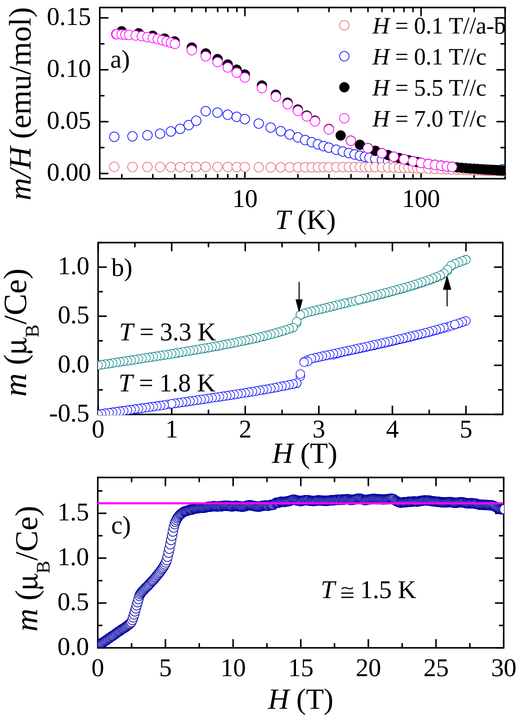

FIG. 3: a) Magnetic moment m of $CeAuSb_2$ divided by the field H, as a function of T for several values of H. b) m of Ce, measured in a SQUID magnetometer, as a function of field H for $T = 3.3$ K and 1.8 K (The vertical scale for the latter is off-set by -0.5). Vertical arrows indicate the fields of both metamagnetic transitions. c) m as a function of H at $T \simeq 1.5$ K measured with a vibrating sample magnetometer.

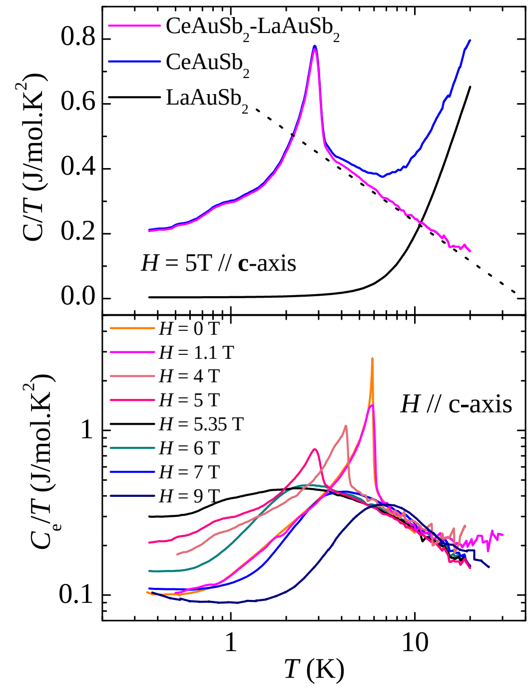

FIG. 4: Upper panel: Heat capacity divided by temperature C/T vs. T for $CeAuSb_2$ with $H = 5$ T applied along the c-axis (blue line), as well as for $LaAuSb_2$ at $H = 0$ T (black line). The difference is the magnetic contribution to the heat capacity C_e/T (in magenta), which shows a $\ln T$-dependence. Lower panel: C_e/T for several magnetic fields. All curves collapse into a single curve at high T and γ ($\gamma = C_e/T$ as $T \to 0$) increases as H approaches H_c.

As seen below, the saturation of the magnetization m suggests that the system is nearly half-metallic for very large H.

The middle panel of Fig. 2 shows the phase diagram resulting from transport and magnetization measurements. The blue triangles indicate the boundary between the PM and the AF phases for $H \parallel$ c-axis, while the green ones define the phase boundary for H in the a-b plane. Notice the remarkable anisotropy. Surprisingly no hysteresis was detected, neither in T nor in H scans, despite the abrupt changes in the slope of $\rho(H)$. This suggests that the transition between the PM and AF phase is a weak first order one or unexpectedly of second-order. The first MM-transition at $H_{\rm MM} \simeq 2.8$ T, which could correspond to a spin-flop transition, is indicated with open circles.

The field dependence of the A and A_3 coefficients (prefactors of the T^2 and T^3 terms) of the resistivity is displayed in the lower panel of Fig. 2. Both coefficients diverge as $H \to H_c$ and the red line is a fit to $C/(H - H_c)$. The same dependence was reported for $YbRh_2Si_2$ [4] and interpreted as a dramatic increase of the quasiparticles linewidth throughout the entire Fermi surface. The A-coefficient also increases at the first MM-transition due to an enhancement in the scattering of the quasiparticles.

The magnetic moment m of a $CeAuSb_2$ single crystal divided by the field is shown in Fig. 3a) as a function of T. For small fields and higher T, m/H follows a Curie-Weiss law, yielding an antiferromagnetic Weiss-temperature $\theta_C = 12.25$ K and an effective moment $p_{eff} \approx 2.26\mu_B$, for the field applied along the c-axis. Notice that the AF transition is observed only for $H \parallel c$, which is the magnetic easy axis. At higher fields, m/H saturates, suggesting the complete polarization of the Ce-spins. In Fig. 3b) both metamagnetic transitions are seen as steps in the magnetization m as a function of H. Both traces contain field-up and down sweeps measured in a SQUID magnetometer. Again, as for the resistivity, no hysteresis is observed. The field of the first MM-transition is only weakly T-dependent. In contrast, as already seen in the resistivity data, the T_N for the second MM-transition (AFM to PM) is markedly field-dependent. Fig. 3c) shows m as a function of H (up to 30 T) at $T = 1.5$ K. Above $H \sim 7$ T, m saturates at $m \simeq 1.65\mu_B$, a value that differs from the free ion moment of Ce due to crystalline electric field effects.

The upper panel of Fig. 4 shows $C(T)/T$ as a function of T for $CeAuSb_2$ at $H = 5$T, and for its isostructural non-magnetic analog $LaAuSb_2$ at $H = 0$T. The

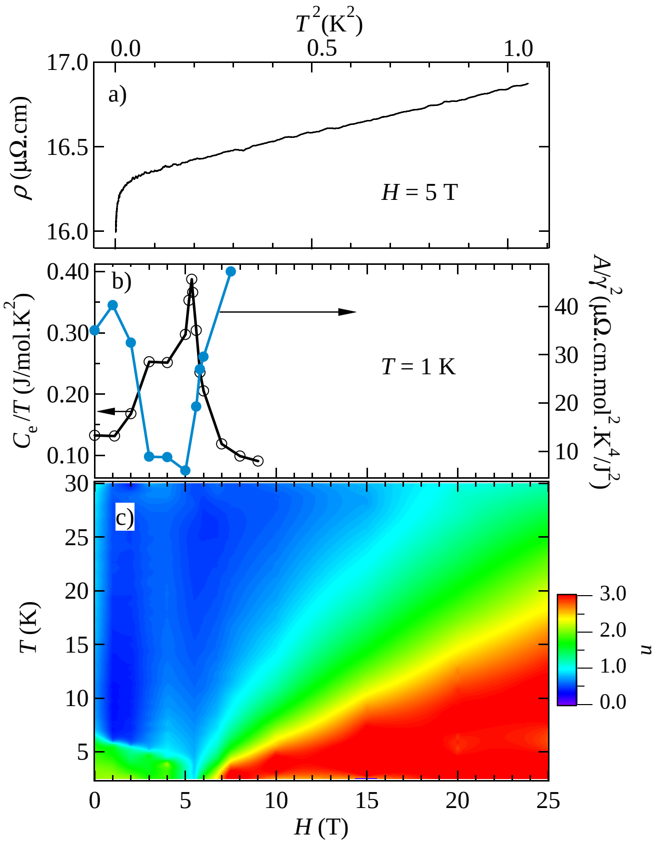

FIG. 5: a) The T^2-dependence of ρ down to $T \simeq 25$ mK for $H = 5$ T $\lesssim H_c$. Notice the pronounced deviation to the T^2 dependence emerging at the lowest Ts. b) The dependence of $C_{e/T}$ for $T = 1$ K (black markers) and the Kadowaki-Woods ratio A/γ^2 (blue markers) as a function of H. There is a sharp increment of C_e/T at H_c. c) The exponent n of $\rho(T)$ in the T - H plane.

large peak for $CeAuSb_2$ signals the AF transition. The subtraction of both curves yields the magnetic contribution to the heat capacity $C_e(T)/T$. For $3 < T < 20$ K, $C_e(T)/T$ displays a $-\ln(T)$ NFL-like dependence. The lower panel of Fig. 4 shows $C_e(T)/T$ as a function of T for several values of H. In the PM phase all curves collapse into a single trace, suggesting that the origin of the $\ln(T)$-dependence is the magnetic pre-critical fluctuations to the AF order. Furthermore, the effective mass of the quasi-particles, as given by γ_e ($C_e(T)/T$ for $T \rightarrow 0$) increases considerably as $H \rightarrow H_c$.

Fig. 5a) shows the T^2-dependence of ρ down to $T \simeq 25$ mK for $H = 5$ T $\lesssim H_c$. There is evidence for a pronounced reduction ρ at the lowest T. In Fig. 5b) we display $C_e(T)/T$ at $T = 1$ K as a function of H by the black (open) markers. This shows that γ dramatically increases at the critical field, but that it remains finite. In other words, the $-\ln(T)$-dependence does not continue to very low T. The blue (closed) represent the Kadowacki-Woods ratio, A/γ^2, as a function of H. The fact that this ratio is not constant indicates that there is more than one energy scale involved.

Fig. 5c) depicts a qualitative sketch of the phase diagram. It shows the dependence of the exponent $n \simeq \partial \ln(\rho(T) - \rho_0)/\partial \ln T$ on H and T. Here different values of ρ_0 were used in the PM and AF phases. The PM phase is indicated by the blue region which is influenced by the QCP leading to the anomalous NFL value $n \lesssim 1$. The FL state (in green) is recovered below the Néel temperature but is gradually suppressed as $H \rightarrow H_c$. As for $YbRh_2Si_2$ [4, 5] a FL-like $n = 2$ exponent is obtained above H_c but only over a limited range of T. A spin-polarized metallic phase (in red) with an anomalous T^3-dependence is observed for $H > H_c$ at low T. Besides $Sr_3Ru_2O_7$ [7], such dependence has also been predicted for half-metallic ferromagnets in terms of a one magnon scattering process [18]. This could be a plausible explanation for our system, which is spin-polarized precisely in the region where $n = 3$ is observed. Although $CeAuSb_2$ is not a FM in zero-field, it could have similar properties to a FM half-metal in a strong magnetic field.

In summary, electrical transport, magnetization and thermal properties indicate that $CeAuSb_2$ displays either a weak first-order or a second-order phase transition from a NFL-like PM metallic phase to a FL phase with long-range AF-order upon cooling at zero field. A magnetic field continuously reduces T_N, which vanishes for a field of $H_c = 5.3 \pm 0.2$ T along the inter-layer direction. The physical properties of the PM phase emerging at H_c are anomalous, i.e., $\rho(T) \propto T^{1/2}$, and $C(T)/T \propto -\ln(T)$, and are a strong indication that H_c corresponds to a field-tuned QCP. In addition, the A and A_3 coefficients of the resistivity diverge as $H \rightarrow H_c$. The consequences of the QCP in $CeAuSb_2$ are different and perhaps more dramatic than for other field-tuned QCP systems. This is the case because the FL phase is not recovered for H and T sufficiently far away from the QCP. The narrow region in Fig. 5 with $n = 2$ should be considered a crossover region and not a FL phase. The field-tuned QCP systems represent a challenge from the theoretical perspective, since the different compounds have some common aspects, but do *not* seem to belong to the same universality class.

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